

Air pollution in crisis: How the Greek economic collapse impacted space-borne formaldehyde levels

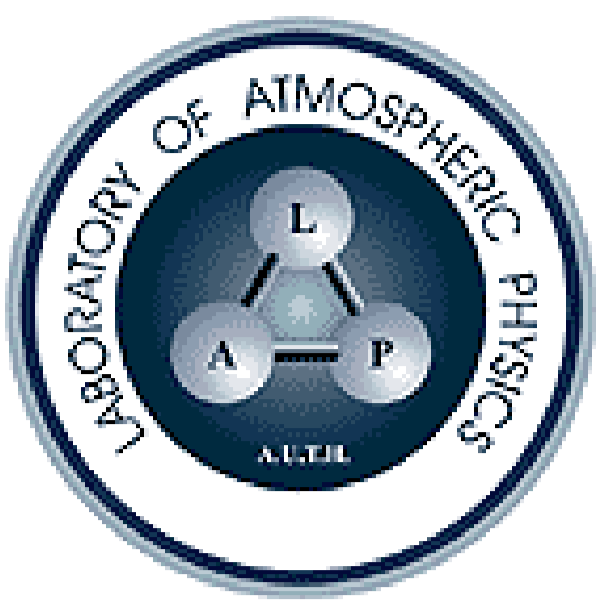
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ABSTRACT

Urban Greek regions (Thessaloniki and Athens) displaying increased HCHO vertical column densities (up to 15×10^{15} molecules/cm²) that are probably linked to anthropogenic emissions (biomass burning and urban pollution) have been identified using satellite observations during wintertime. Based on a continuous six-year record (2008-2013) of HCHO columns inferred from GOME-2/MetOp-A and OMI/Aura satellite measurements, we explore how the urban air quality is affected during the winter months due to the financial problems that plague Greece. In particular, a larger positive change of approximately $6.5 \pm 0.1\%$ has emerged since 2009 from the morning HCHO enhancements observed by GOME-2 than by OMI (about $2.0 \pm 0.1\%$). We further compared mean monthly HCHO observations with simulated HCHO columns from the CAMx chemical transport model as well as with its emissions' seasonal variability in order to study the agreement on the annual cycle. The winter observed HCHO year-to-year variability was explored and compared to the surface temperatures in order to explore the origin of the winter HCHO enhancements that are not being captured by the model. A clear anti-correlation was found since the beginning of the economic recession in the country and especially over Thessaloniki. The intensive and increased use of woods and pellets for domestic heating due to the inhabitants' decreased income and the exorbitant tax increase on fossil fuel energy appear to be the dominant reasons of the wintertime increase in anthropogenic HCHO sources.

SATELLITE OBSERVATIONS

The **second Global Ozone Monitoring Experiment (GOME-2)** UV spectrometer flies on EUMETSAT's MetOp-A spacecraft since October 2006 in a sun synchronous near-polar orbit, with a local Equator crossing time of **09:30** (descending node). GOME-2 is described in details in Callies et al. (2000) and Hassinen et al. (2016).

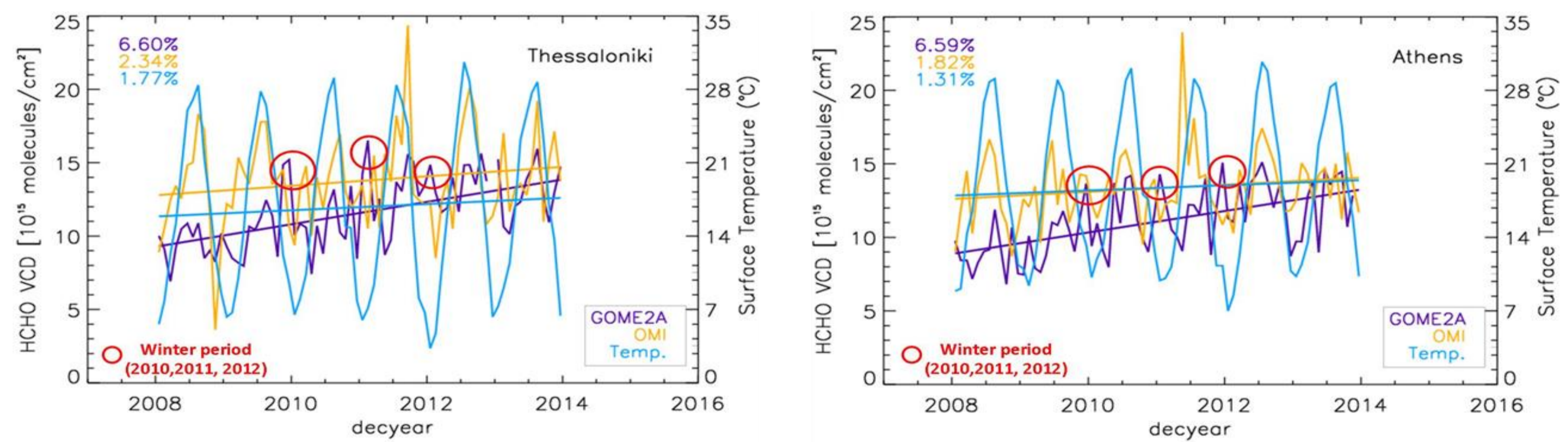
The **Ozone Monitoring Instrument (OMI)** was launched onboard the NASA's EOS-Aura satellite in July 2004 into a sun-synchronous polar orbit crossing the Equator around **13:30** LT (in ascending mode). OMI is described in details in Levelt et al. (2006).

We used the **new version (v14) of the BIRA/IASB algorithm** for the retrieval of HCHO columns from GOME-2 and OMI available at <http://h2co.aeronomie.be/>. (De Smedt et al., 2015). HCHO slant columns are retrieved from a fitting window of **328.5–346 nm** using **DOAS** and the HCHO absorption cross-sections of Meller and Moortgat [2000] as described in De Smedt et al. 2012.

METHODOLOGY

We used quality criteria for some of the important parameters such as: effective **cloud fractions less than 0.4**, **solar zenith angles less than 70°** and **individual vertical column errors less than three times the column** (De Smedt et al. 2015). **As for OMI only the unaffected pixels of OMI row anomalies were used in our analysis.** We also estimated the total error on column averages combining the random and systematic contributions (De Smedt et al. 2014). We applied the **averaging kernels** (given in GOME2A & OMI product) to the model. The HCHO vertical column densities were extracted by integrating the produced CAMx profiles (17 layers) from the surface up to about 10km. **The mean monthly total error ranges from about 3.0-7.0 and from 7.0-11.0 $\times 10^{15}$ molecules/cm² for GOME-2 and OMI respectively.** The satellite observations were averaged within 50km around each location and compared with **model results sampled at the time of the satellite overpass.**

TIMESERIES OF HCHO RETRIEVALS



The above figures depict the monthly mean time series of HCHO observations and their corresponding linear trend over Thessaloniki (left) and Athens (right) as observed by GOME-2 (purple line) and OMI (yellow line). The light blue line represents the surface temperature derived from <http://data.giss.nasa.gov/gistemp/> [GISTEMP Team, 2016] [Hansen et al., 2010]. There is a **positive trend** for HCHO over the two urban Greek regions which is **higher for GOME-2 (~6.5±0.12%) than OMI (~2±0.16%)**. The annual change is positive for GOME-2A and OMI, the differences in absolute values are partly due to the combined effect of their different overpass time and the short HCHO lifetime and their trends are within their combined uncertainties. Although the satellite HCHO observations and the surface temperature are usually well correlated, there are some cold periods (in 2010, 2011 and 2012) that they are unexpectedly anticorrelated.

CONCLUSIONS

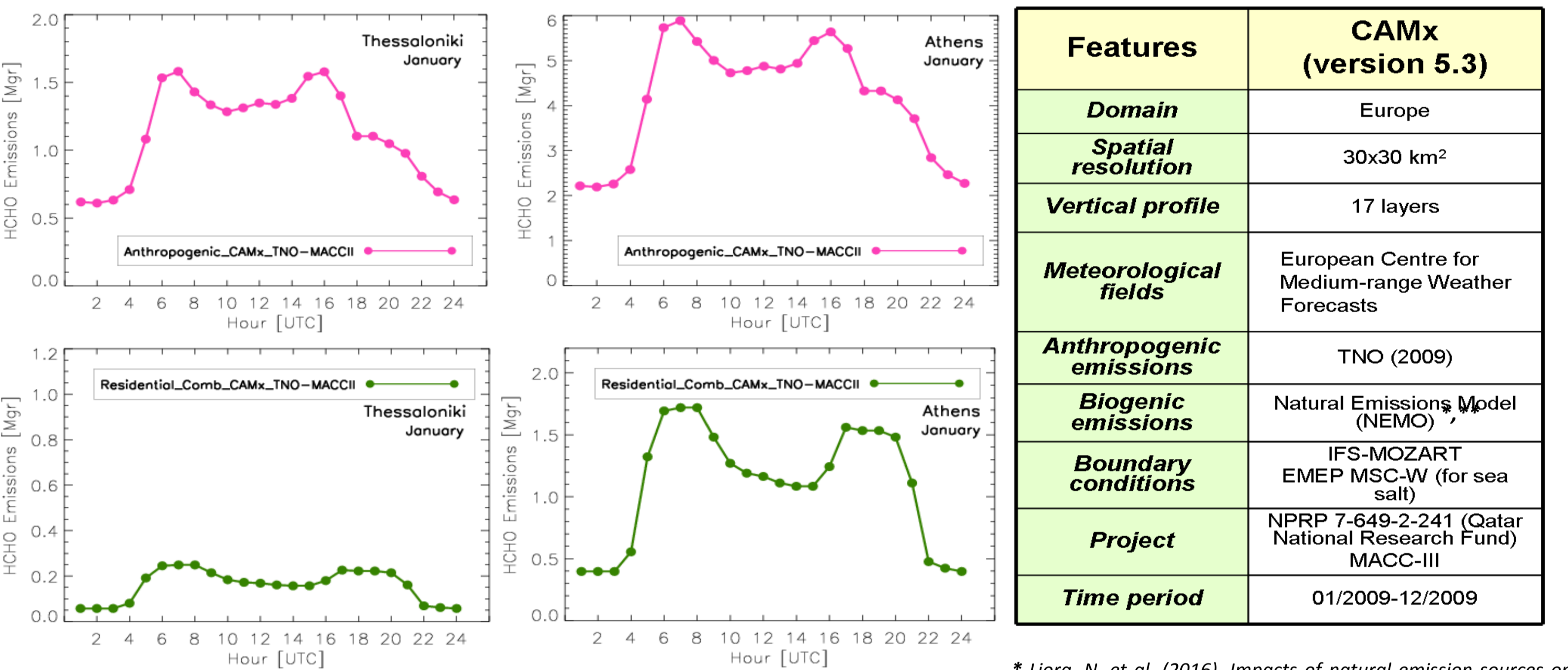
In this study significant HCHO column enhancements were observed from space over Thessaloniki and Athens between 2008 and 2013 giving a **positive trend of ~6.5%** in the mid-morning (**from GOME2A observations**) and **~2.0%** in the early afternoon (**from OMI observations**) with **1-sigma uncertainty of around 0.1%** for both instruments. The differences between mid-morning and early afternoon trends are probably attributed to the different instruments' features. In this study **the increased use of woods and pellets for residential heating is reflected in higher HCHO levels in January values of 2011, 2012 and 2013 observed both by GOME2A and OMI mostly over Thessaloniki.** 2011 seems to be the year with not only the maximum HCHO concentrations but also with GOME2A observations greater than the OMI ones. Since 2009 in Athens and 2010 in Thessaloniki **the GOME2A HCHO VCDs started increasing and being anti-correlated with the surface temperature.** The above findings temporally coincide with the outbreak of the financial recession in Greece and thus denote a **possibly clear relationship between the enhanced HCHO levels in the cold winter mornings and the extensive biomass burning activities (firewood, pellets, alternative fuels)** in its urban atmosphere. Such observed **HCHO enhancements are not captured by the anthropogenic emission inventory used in CAMx model**, especially for Thessaloniki. It has been shown that satellite observations are an important means in order to constrain the emission budget of HCHO in urban atmospheres with similar air pollution problems in wintertime.

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CAMx MODEL

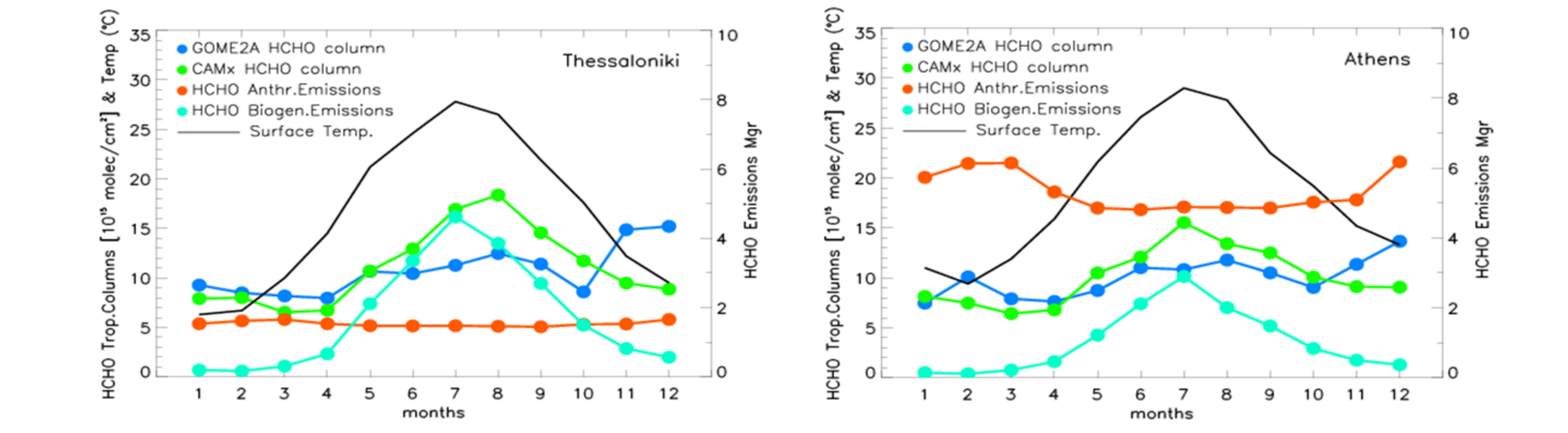
The modeling system used in the present study, consisted of the Weather Research and Forecasting model (WRF v. 3.5.1; Skamarock et al., 2008) and the three-dimensional Comprehensive Air Quality Model with extensions (CAMx v.5.3; ENVIRON, 2010).



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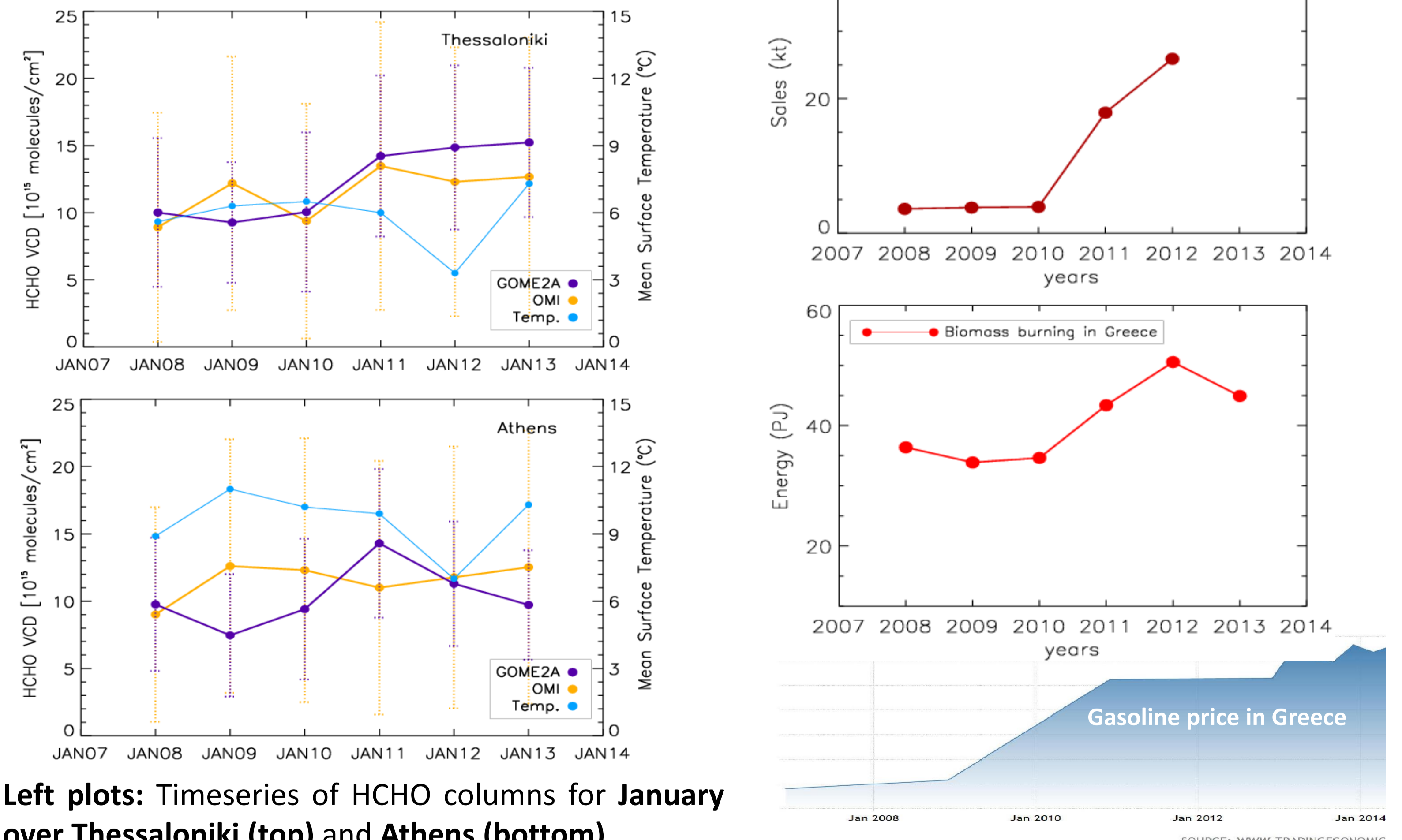
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SEASONAL VARIATION OF HCHO COLUMNS



The above two Figures illustrate **the annual variability of the HCHO columns as observed by GOME2A (deep blue dots) and simulated by CAMx (green dots) in 10^{15} molecules/cm², the surface temperature (black line) in °C (left y-axis) and the anthropogenic (orange dots) and biogenic (light blue dots) HCHO emissions in Mg (right y-axis) over Thessaloniki (left) and Athens (right) for the year 2009.** The CAMx simulations perform a clear seasonal pattern similar to the seasonality of the biogenic emissions and the surface temperature peaking in summer, which is qualitatively in phase with the main crop growing season (April to September) over both cities. GOME-2 HCHO columns provide values in the cold months (15×10^{15} molecules/cm²) that are comparable and in some cases greater than those during the hot ones which might denote the emergence of winter anthropogenic HCHO primary emissions. These winter enhancements are not captured well by the CAMx anthropogenic emission inventory especially over Thessaloniki.

WINTER HCHO EMISSIONS HOTSPOTS



Left plots: Timeseries of HCHO columns for **January over Thessaloniki (top) and Athens (bottom).**

Right plots: (top) the retail sales of wood and pellet in the greater area of Thessaloniki since the beginning of the economic recession [Slini et al., 2015], (middle) the energy consumption for residential heating by biomass burning in Greece [Odyssee-Mure project, 2014], (bottom) timeseries of the gasoline price in Greece.

In winter plots the **GOME-2 HCHO values (purple line) were increased since 2010 whereas the surface ature were decreased (blue line) over both urban regions.** The positive trend of GOME-2 HCHO values are **consistent with the increased gasoline price in Greece since 2010 that led to enhanced use of wood in the area.**

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